

Gradient flow for controlling quantum ensemble

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Abstract—We propose in this paper a gradient-type dynamical system to solve the problem of maximizing quantum observables for finite dimensional closed quantum ensembles governed by the controlled Liouville-von Neumann equation. The asymptotic behavior is analyzed: we show that under the regularity assumption on the controls the dynamical system almost always converges to a solution of the maximization problem; we also detail the difficulties related to the occurrence of singular controls.

I. INTRODUCTION

Quantum control is concerned with actively manipulating physical and chemical processes on the atomic or molecular scale where quantum mechanics is the rule. The origin of quantum control goes back to the early attempts to use lasers for selectively breaking molecular bonds, and several approaches using quantum interference, adiabatic passage, pump-dump control etc. have been proposed since 1970's. For the historical development of quantum control, the state of the art from both theoretical and experimental points of view, and open research directions, see for instance the recent review paper [7]. An overview on control techniques applied to manipulating quantum systems is also given in [11]. More detailed treatment from a control theoretical point of view can be found in [9]. Among existing methods for controlling quantum systems, optimal control theory plays a major role. The key point is to develop control strategies in a constructive way such that a certain performance index, or cost functional is optimized under the constraints imposed by realistic experimental conditions. Three classes of problems - state transition, observable maximization, and unitary transformation- have been attracting the most attention in the community [7], [23]. The performance indices in these problems only depend on the final states of the corresponding quantum systems, although in full generality time or energy consumption could be taken into account as well (see for instance [17], [20], [6]). Moreover, these performance indices can also be used as Lyapunov functions in closed-loop feedback designs for stabilization or trajectory tracking, see [29], [22], [21], [4], [28] and references therein. In this paper, we only consider the problem of maximizing quantum observables for closed quantum systems, an open loop strategy will be proposed. The analysis also extends to state-transition and unitary transformation problems.

For a closed n -level quantum system, the evolution of its density matrix $\rho(t)$ under the dipole moment approximation is described by the following time-varying Liouville-von Neumann equation:

$$\begin{cases} \dot{\rho}(t) &= [H_0 + u(t)H_1, \rho(t)], & t \in [0, T], \\ \rho(0) &= \rho_0, \end{cases} \quad (1)$$

where ρ_0 is the initial density matrix which is assumed to be Hermitian, the traceless skew-Hermitian matrices H_0 and H_1 are respectively the free Hamiltonian of the system and the dipole moment. The vector space of traceless skew-Hermitian matrices will be denoted by $\mathfrak{su}(n)$. We assume that the admissible controls u are elements of $\mathcal{H} := L^2([0, T], \mathbb{R})$. This corresponds to the ideal case where the intensity of the external field $u(\cdot)$ is not constrained. The vector space \mathcal{H} equipped with the standard inner product $(u, v)_{\mathcal{H}} = \int_0^T u(t)v(t)dt$, for $(u, v) \in \mathcal{H} \times \mathcal{H}$, is a Hilbert space. The corresponding norm will be denoted by $\|\cdot\|_{\mathcal{H}}$.

It is well-known that the solution of (1) is given by

$$\rho(t) = U(t)\rho_0U(t)^\dagger, \quad (2)$$

where the propagator $U(\cdot)$ satisfies

$$\begin{cases} \dot{U}(t) &= (H_0 + u(t)H_1)U(t), & t \in [0, T], \\ U(0) &= \text{Id}. \end{cases} \quad (3)$$

Since H_0 and H_1 belong to $\mathfrak{su}(n)$, $U(\cdot)$ is a curve in the special unitary group $\text{SU}(n)$. Recall that $\text{SU}(n)$ is a compact Lie group and its Lie algebra is $\mathfrak{su}(n)$. Eq. (2) implies that ρ evolves in a subset of the unitary orbit of ρ_0 defined by

$$\mathcal{O}(\rho_0) := \{U\rho_0U^\dagger, U \in \text{SU}(n)\}.$$

We assume from now that the system (3) is controllable, then the state space of ρ is equal to $\mathcal{O}(\rho_0)$ and the system (1) is controllable in the sense that all points of $\mathcal{O}(\rho_0)$ can be reached from ρ_0 by choosing suitable controls.

Remark I.1. For T large enough, a necessary and sufficient condition for (3) to be controllable is that the Lie algebra generated by H_0 and H_1 is equal to $\mathfrak{su}(n)$. This is a consequence of the controllability results on general Lie groups obtained by Jurdjevic and Sussmann in [16]. See [24], [12], [25], or [9, Ch. 3] for controllability of quantum systems. We also note that the set of pairs (H_0, H_1) such that H_0 and H_1 generate $\mathfrak{su}(n)$ is open and dense in $\mathfrak{su}(n) \times \mathfrak{su}(n)$ (cf. [15, Th. 12, Ch. 6, p 188]).

Define the end-point map for (1) as

$$\text{End}_{\rho_0}(\cdot) : \begin{array}{ccc} \mathcal{H} & \mapsto & \mathcal{O}(\rho_0) \\ u & \mapsto & \rho(T) \end{array}.$$

In this paper, we are interested in the following maximization problem:

Problem 1. Let θ be a Hermitian matrix. Find $u_{\max} \in \mathcal{H}$ maximizing the cost function

$$\mathcal{J}(u) := \text{Re tr}(\text{End}_{\rho_0}(u)\theta), \quad \text{for } u \in \mathcal{H}. \quad (4)$$

Remark I.2. θ represents an observable for the quantum

system and $\text{tr}(\rho(T)\theta)$ is the average of different possible results given by the measurement of θ at time T (cf. [8, Chap 3-E]). **Problem 1** consists in finding a control field u maximizing this average.

This problem is closely related to the two following problems.

Problem 2. Let θ be a Hermitian matrix. Find $\rho_{\max} \in \mathcal{O}(\rho_0)$ maximizing the cost function

$$J(\rho) := \text{Re } \text{tr}(\rho\theta), \quad \text{for } \rho \in \mathcal{O}(\rho_0). \quad (5)$$

Problem 3. Given an arbitrary target state $\rho^{\text{final}} \in \mathcal{O}(\rho_0)$, find $u^{\text{final}} \in \mathcal{H}$ such that

$$\text{End}_{\rho_0}(u^{\text{final}}) = \rho^{\text{final}}. \quad (6)$$

Remark I.3. The compactness of $\mathcal{O}(\rho_0)$ guarantees the existence of solutions for **Problem 2**, which in turn implies, together with the controllability assumption, the existence of solutions for **Problem 1**.

Remark I.4. If we are able to find ρ_{\max} a solution to **Problem 2**, then **Problem 1** is equivalent to **Problem 3** with target state equal to ρ_{\max} . We also note that u is a solution to **Problem 1** if and only if $\text{End}_{\rho_0}(u)$ is a solution to **Problem 2**.

We discuss in this paper a gradient-type dynamical system to solve **Problem 1**. The method is well-known in the quantum chemistry and NMR (Nuclear Magnetic Resonance) communities (see for example [14], [7], [18]). We give here a rigorous mathematical formulation of this method as well as analysis on its asymptotic behavior. We also formulate some open questions related to the presence of singular controls for (1). The paper is organized as follows. We recall in Section II classical results on the geometry of the unitary orbit $\mathcal{O}(\rho_0)$ and derive some computational lemmas related to the end-point map. The main results of this paper concerning the asymptotic behavior of the dynamical system are presented in Section III. Finally, concluding remarks are formulated in Section IV and Appendix deals with a technical proof.

II. PRELIMINARY RESULTS

A. Geometry of the unitary orbit

We summarize in this paragraph some results on the geometry of the unitary orbit $\mathcal{O}(\rho_0)$. The key point is to define a suitable Riemannian metric on $\mathcal{O}(\rho_0)$. Add references. The presentation here follows [26, Section 3.4.4].

Recall that $\mathcal{O}(\rho_0)$ is a compact connected submanifold of $\mathbb{C}^{n \times n}$ isomorphic to the quotient space $\text{SU}(n)/\mathbf{H}$, where

$$\mathbf{H} := \{U \in \text{SU}(n), U\rho_0 U^\dagger = \rho_0\}$$

denotes the stabilizer group of ρ_0 . We have

$$\dim \mathcal{O}(\rho_0) = n^2 - 1 - \dim \mathbf{H} := N. \quad (7)$$

The tangent space of $\mathcal{O}(\rho_0)$ at $\rho = \text{Ad}_U \rho_0 := U\rho_0 U^\dagger$ is given by

$$T_\rho \mathcal{O}(\rho_0) = \text{ad}_\rho \mathfrak{su}(n) = \{\text{ad}_\rho \Omega, \Omega \in \mathfrak{su}(n)\}, \quad (8)$$

with $\text{ad}_\rho \Omega := [\rho, \Omega] := \rho\Omega - \Omega\rho$.

Remark II.1. Since the adjoint map $\text{Ad}_U : \Omega \mapsto \text{Ad}_U \Omega$ defines an automorphism on $\mathfrak{su}(n)$, the tangent space $T_\rho \mathcal{O}(\rho_0)$ is also equal to $\{\text{ad}_\rho \text{Ad}_U \Omega, \Omega \in \mathfrak{su}(n)\}$.

In order to define the gradient of the cost function J , we first need to equip $T_\rho \mathcal{O}(\rho_0)$ with a scalar product. Note that the kernel of $\text{ad}_\rho : \mathfrak{su}(n) \mapsto \mathbb{C}^{n \times n}$ is given by

$$\mathfrak{h} := \{\Omega \in \mathfrak{su}(n), [\rho_0, \Omega] = 0\}$$

and forms the Lie subalgebra to \mathbf{H} . By the standard Hilbert-Schmidt scalar product $(\Omega_1, \Omega_2) \mapsto \text{tr}(\Omega_1^\dagger \Omega_2)$ on $\mathfrak{su}(n)$ one can define the ortho-complement of \mathfrak{h} as

$$\mathfrak{p} := \{\Omega_1 \in \mathfrak{su}(n), \text{tr}(\Omega_1^\dagger \Omega_2) = 0, \text{ for all } \Omega_2 \in \mathfrak{h}\}.$$

This induces a unique decomposition of any skew-Hermitian matrix $\Omega = \Omega^{\mathfrak{h}} + \Omega^{\mathfrak{p}}$ with $\Omega^{\mathfrak{h}} \in \mathfrak{h}$ and $\Omega^{\mathfrak{p}} \in \mathfrak{p}$.

Definition II.1. For $\rho = \text{Ad}_U \rho_0$ with $U \in \text{SU}(n)$, we define a scalar product $\langle \cdot, \cdot \rangle_\rho$ on $T_\rho \mathcal{O}(\rho_0)$ by

$$\langle \text{ad}_\rho(\text{Ad}_U \Omega_1), \text{ad}_\rho(\text{Ad}_U \Omega_2) \rangle_\rho := \text{tr}(\Omega_1^{\mathfrak{p}} \Omega_2^{\mathfrak{p}}), \quad (9)$$

which is equivalent to

$$\langle \text{ad}_\rho \Omega_1, \text{ad}_\rho \Omega_2 \rangle_\rho := \text{tr}(\Omega_1^{\mathfrak{p}_\rho} \Omega_2^{\mathfrak{p}_\rho}) \quad (10)$$

with $\mathfrak{p}_\rho := \text{Ad}_U \mathfrak{p}$.

A fundamental property of the Riemannian metric defined above is it is $\text{Ad}_{\text{SU}(n)}$ -invariant, i.e., $\forall \xi, \eta \in T_\rho \mathcal{O}(\rho_0)$, and $\forall U \in \text{SU}(n)$,

$$\langle \xi, \eta \rangle_\rho = \langle \text{Ad}_U \xi, \text{Ad}_U \eta \rangle_{\text{Ad}_U \rho}. \quad (11)$$

For later use, we recall the following result.

Proposition II.1 (Theorem 3.16 [26]). Let J be the cost function considered in **Problem 2** and $\rho \in \mathcal{O}(\rho_0)$. Then, the gradient of J at ρ with respect to the Riemannian metric defined by Eq. (9) is given by

$$\nabla J(\rho) = [\rho, [\rho, \theta]].$$

Furthermore, $\rho_c \in \mathcal{O}(\rho_0)$ is a critical point of J if and only if

$$[\rho_c, \theta] = 0.$$

Remark II.2. Let $dJ(\rho)$ be the differential of J at ρ . Then, by definition, we have

$$\forall \eta \in T_\rho \mathcal{O}(\rho_0), dJ(\rho)\eta = \langle \nabla J(\rho), \eta \rangle_\rho.$$

We note that the expression of the gradient depends on the metric chosen for $T_\rho \mathcal{O}(\rho_0)$. One can choose metrics other than the one defined by (9), for example, the induced Riemannian metric if we consider $\mathcal{O}(\rho_0)$ as a submanifold embedded in $\mathbb{C}^{n \times n}$. However, the invariant metric defined above gives a simple expression of ∇J .

In order to simplify the discussion, we assume that

(H1) the initial density matrix ρ_0 and the observable O both have simple eigenvalues.

The following result is a direct consequence of Proposition II.1 and **(H1)**.

Corollary II.2. Under (H1), J has $M := n!$ isolated critical points in $\mathcal{O}(\rho_0)$.

Let $\{\rho_i\}_{i=1,\dots,M}$ be the critical points of J such that $J(\rho_1) \leq \dots \leq J(\rho_M)$. For $i = 1, \dots, M$, let $\nabla^2 J(\rho_i)$ be the Hessian of J at ρ_i .

Lemma II.3. Under (H1), for $i = 1, \dots, M$, $\nabla^2 J(\rho_i)$ is non-degenerate. Moreover, $\nabla^2 J(\rho_1)$ is positive definite, $\nabla^2 J(\rho_M)$ is negative definite, and $\nabla^2 J(\rho_i)$ is not definite for $i = 2, \dots, M-1$.

Lemma II.3 states that J only has one minimum and one maximum, all other critical points are saddles. The proof is a straightforward adaptation of the one for [13, Th. 1.3, p 52]. See also [26, Cor. 3.8] and its proof.

B. Differential of the end-point map and its adjoint operator

The end-point map $\text{End}_{\rho_0}(\cdot)$ is C^∞ (in fact analytical in our case). For $u \in \mathcal{H}$, the first derivative of End_{ρ_0} at u is given by

$$\begin{aligned} d\text{End}_{\rho_0}(u) : \mathcal{H} &\mapsto T_{\text{End}_{\rho_0}(u)}\mathcal{O}(\rho_0) \\ v &\mapsto d\text{End}_{\rho_0}(u)v = y_v(T), \end{aligned} \quad (12)$$

where, for every $v \in \mathcal{H}$, $y_v : [0, T] \mapsto T\mathcal{O}(\rho_0)$ is the solution of the variational equation

$$\begin{cases} \dot{y}(t) &= [H_0 + u(t)H_1, y(t)] + v(t)[H_1, \rho(t)], \quad t \in [0, T], \\ y(0) &= 0, \end{cases} \quad (13)$$

with $\rho(\cdot)$ denoting the solution of (1) associated with the control u . The following computational lemma is obtained by variation of constants.

Lemma II.4. If $U(\cdot) : [0, T] \mapsto \text{SU}(n)$ satisfies

$$\begin{cases} \dot{U}(t) &= (H_0 + u(t)H_1)U(t), \quad t \in [0, T], \\ U(0) &= \text{Id}, \end{cases}$$

then, $y_v(\cdot) : [0, T] \mapsto T\mathcal{O}(\rho_0)$ given by

$$y_v(t) = U(t) \int_0^t [U^\dagger(s)H_1U(s), \rho_0]v(s)ds U(t)^\dagger \quad (14)$$

is the solution of (13).

Corollary II.5. There exists a constant $\tilde{C} > 0$ depending on ρ_0, H_1, T such that for all $u \in \mathcal{H}$, we have

$$\|d\text{End}_{\rho_0}(u)v\| \leq \tilde{C}\|v\|_{\mathcal{H}}, \quad \forall v \in \mathcal{H}. \quad (15)$$

Proof of Corollary II.5: It suffices to note that there exists a constant $C_1 > 0$ such that

$$\|z(t)\| \leq C_1\|v\|_{\mathcal{H}}, \quad \text{for all } t \in [0, T], \text{ and } u \in \mathcal{H},$$

where $z(t) = \int_0^t [U^\dagger(s)H_1U(s), \rho_0]v(s)ds$. ■

Definition II.2. A control $u \in \mathcal{H}$ is called *regular* if the rank of $d\text{End}_{\rho_0}(u)$ is equal to the dimension of the state space $\mathcal{O}(\rho_0)$. The corresponding trajectory is called *regular trajectory*.

Definition II.3. A control $u \in \mathcal{H}$ is called *singular* if the rank of $d\text{End}_{\rho_0}(u)$ is smaller than the dimension of the state space $\mathcal{O}(\rho_0)$. The corresponding trajectory is called *singular trajectory*. The *co-rank* of a singular control u is defined as equal to

$$\dim \mathcal{O}(\rho_0) - \text{rank}(d\text{End}_{\rho_0}(u)).$$

Remark II.3. The notion of regular and singular controls will play a crucial role in the convergence analysis of the gradient flow, see Section III for more detail.

Definition II.4. Given $u \in \mathcal{H}$, let $\rho(\cdot)$ be the solution of

$$\begin{cases} \dot{\rho}(t) &= [H_0 + u(t)H_1, \rho(t)], \quad t \in [0, T], \\ \rho(0) &= \rho_0. \end{cases} \quad (16)$$

The *adjoint equation* along $\rho(\cdot)$ is defined by

$$\begin{cases} \dot{q}(t) &= [H_0 + u(t)H_1, q(t)], \quad t \in [0, T], \\ q(T) &= q_T, \end{cases} \quad (17)$$

for some $q_T \in T_{\rho(T)}\mathcal{O}(\rho_0)$. The solution $q(\cdot)$ of Eq. (17) is called *adjoint vector*. The corresponding *switching function* $\Phi_{\rho_0, q_T}(\cdot)$ is defined by

$$\Phi_{\rho_0, q_T}(t) := \langle q(t), [H_1, \rho(t)] \rangle_{\rho(t)}, \quad (18)$$

where the Riemannian metric $\langle \cdot, \cdot \rangle_{\rho(t)}$ is chosen to be the one given in Definition II.1.

Lemma II.6. For $q_T \in T_{\rho(T)}\mathcal{O}(\rho_0)$ and $v \in \mathcal{H}$, we have

$$\langle q_T, d\text{End}_{\rho_0}(u)v \rangle_{\rho(T)} = \langle \Phi_{\rho_0, q_T}, v \rangle_{\mathcal{H}}. \quad (19)$$

Proof of Lemma II.6:

$$\begin{aligned} &\langle q_T, d\text{End}_{\rho_0}(u)v \rangle_{\rho(T)} \\ &= \langle q_T, U(T) \int_0^T [U(s)^\dagger H_1 U(s), \rho_0]v(s)ds U(T)^\dagger \rangle_{\rho(T)} \\ &= \int_0^T \langle q_T, U(T)[U^\dagger(s)H_1U(s), \rho_0]U^\dagger(T) \rangle_{\rho(T)}v(s)ds. \end{aligned}$$

Since the Riemannian metric $\langle \cdot, \cdot \rangle_{\rho(t)}$ is Ad_{SU} -invariant, we have

$$\begin{aligned} \Phi_{\rho_0, q_T}(t) &= \langle q(t), [H_1, \rho(t)] \rangle_{\rho(t)} \\ &= \langle \text{Ad}_{U(T-t)}q(T), \text{Ad}_{U(T-t)}[H_1, \rho(t)] \rangle_{\text{Ad}_{U(T-t)}\rho(t)} \\ &= \langle q_T, U(T-t)[H_1, \rho(t)]U(T-t)^\dagger \rangle_{\rho(T)} \\ &= \langle q_T, U(T)[U^\dagger(t)H_1U(t), \rho_0]U(T)^\dagger \rangle_{\rho(T)}. \end{aligned} \quad (20)$$

This implies $\langle q_T, d\text{End}_{\rho_0}(u)v \rangle_{\rho(T)} = \langle \Phi_{\rho_0, q_T}, v \rangle_{\mathcal{H}}$. ■

Definition II.5. The adjoint operator $d\text{End}_{\rho_0}^*(u)$ of $d\text{End}_{\rho_0}(u)$ is defined as the unique operator satisfying

$$\langle z, d\text{End}_{\rho_0}(u)v \rangle_{\text{End}_{\rho_0}(u)} = \langle d\text{End}_{\rho_0}^*(u)z, v \rangle_{\mathcal{H}},$$

for all $z \in T_{\text{End}_{\rho_0}(u)}\mathcal{O}(\rho_0)$ and $v \in \mathcal{H}$.

From Lemma II.6 and Definition II.5, we immediately get the following corollary.

Corollary II.7. For $z \in T_{\text{End}_{\rho_0}(u)}\mathcal{O}(\rho_0)$, we have

$$d\text{End}_{\rho_0}^*(u)z = \Phi_{\rho_0, z}. \quad (21)$$

Definition II.6. For $u \in \mathcal{H}$, the non-negative symmetric matrix called *controllability Gramian* of (13) is defined by

$$G(u) := d\text{End}_{\rho_0}(u)d\text{End}_{\rho_0}^*(u).$$

The following fundamental property holds.

Proposition II.8. For all $z \in T_{\text{End}_{\rho_0}(u)}\mathcal{O}(\rho_0)$, we have

$$\langle z, G(u)z \rangle_{\text{End}_{\rho_0}(u)} = \|d\text{End}_{\rho_0}^*(u)z\|_{\mathcal{H}}^2 = \|\Phi_{\rho_0, z}\|_{\mathcal{H}}^2,$$

and

$$\text{rank } d\text{End}_{\rho_0}(u) = \dim \mathcal{O}(\rho_0) \iff G(u) \text{ is positive definite.}$$

For later use, we finish this section by giving the second derivative of $\text{End}_{\rho_0}(\cdot)$ at u in the direction $v \in \mathcal{H}$.

$$d^2\text{End}_{\rho_0}(u) : \begin{array}{ccc} \mathcal{H} & \mapsto & T_{\text{End}_{\rho_0}(u)}\mathcal{O}(\rho_0) \\ v & \mapsto & d^2\text{End}_{\rho_0}(u)(v, v) = r_v(T) \end{array}, \quad (22)$$

where, for every $v \in \mathcal{H}$, $r_v : [0, T] \mapsto T\mathcal{O}(\rho_0)$ is the solution of the second variational equation

$$\begin{cases} \dot{r}(t) &= [H_0 + u(t)H_1, r(t)] + v(t)[H_1, y_v(t)], \quad t \in [0, T], \\ r(0) &= 0, \end{cases} \quad (23)$$

with $y_v(\cdot)$ denoting the solution of the first variational equation (13).

The following lemma is straightforward.

Lemma II.9. If $U(\cdot) : [0, T] \mapsto \text{SU}(n)$ satisfies

$$\begin{cases} \dot{U}(t) &= (H_0 + u(t)H_1)U(t), \quad t \in [0, T], \\ U(0) &= \text{Id}, \end{cases}$$

then, $r_v(\cdot) : [0, T] \mapsto T\mathcal{O}(\rho_0)$ given by

$$r_v(t) = U(t) \int_0^t [U^\dagger(s)H_1U(s), z_v(s)]v(s)ds U(t)^\dagger, \quad (24)$$

with $z_v(\cdot) := \int_0^\cdot [U^\dagger(s)H_1U(s), \rho_0]v(s)ds$ is the solution of (23).

Corollary II.10. There exists a constant $\tilde{C} > 0$ depending on ρ_0, H_1, T such that for all $u \in \mathcal{H}$, we have

$$\|d^2\text{End}_{\rho_0}(u)(v, v)\| \leq \tilde{C}\|v\|_{\mathcal{H}}^2, \quad \forall v \in \mathcal{H}. \quad (25)$$

III. GRADIENT FLOW IN \mathcal{H}

A natural idea to tackle **Problem 1**, which is an optimization problem in the infinite dimensional control space \mathcal{H} , is to follow the gradient of \mathcal{J} as an ascent direction in order to increase \mathcal{J} . The purpose of this section is to present in a rigorous way a gradient-type algorithm widely used in the quantum chemistry and NMR communities, see for example [14], [7], [18].

A. Description of the method and some general properties

We first compute the gradient of \mathcal{J} . Note that $\mathcal{J}(u) = J(\text{End}_{\rho_0}(u))$.

Lemma III.1. For $u \in \mathcal{H}$, we have

$$\nabla \mathcal{J}(u) = d\text{End}_{\rho_0}^*(u)\nabla J(\text{End}_{\rho_0}(u)). \quad (26)$$

Proof of Lemma III.1: Given $u \in \mathcal{H}$, for any $v \in \mathcal{H}$, we have

$$\begin{aligned} d\mathcal{J}(u)v &= dJ(\text{End}_{\rho_0}(u))d\text{End}_{\rho_0}(u)v \\ &= \langle \nabla J(\text{End}_{\rho_0}(u)), d\text{End}_{\rho_0}(u)v \rangle_{\text{End}_{\rho_0}(u)} \\ &= (d\text{End}_{\rho_0}^*(u)\nabla J(\text{End}_{\rho_0}(u)), v)_{\mathcal{H}}. \end{aligned}$$

By definition, we have

$$\nabla \mathcal{J}(u) = d\text{End}_{\rho_0}^*(u)\nabla J(\text{End}_{\rho_0}(u)).$$

■

Algorithm 1 Gradient Flow

- (i) Choose an arbitrary control $u_0 \in \mathcal{H}$.
- (ii) Solve the following initial value problem

$$\begin{cases} \frac{d\Pi}{ds}(s) &= \nabla \mathcal{J}(\Pi(s)) \\ \Pi(0) &= u_0 \end{cases}, \quad (27)$$

or more precisely,

$$\begin{cases} \frac{d\Pi}{ds}(s) &= d\text{End}_{\rho_0}^*(\Pi(s))\nabla J(\text{End}_{\rho_0}(\Pi(s))) \\ \Pi(0) &= u_0 \end{cases}. \quad (28)$$

Before giving some preliminary analysis on the algorithm, we first explain how to compute the right-hand side of Eq. (28).

Lemma III.2. For $u \in \mathcal{H}$, we have

$$\begin{aligned} d\text{End}_{\rho_0}^*(u)\nabla J(\text{End}_{\rho_0}(u)) \\ = -\text{tr}([\rho_0, U(T)^\dagger \theta U(T)]U^\dagger(t)H_1U(t)), \end{aligned}$$

where $U(\cdot)$ satisfies

$$\begin{cases} \dot{U}(t) &= (H_0 + u(t)H_1)U(t), \quad t \in [0, 1], \\ U(0) &= \text{Id}. \end{cases}$$

Proof of Lemma III.2: By Corollary II.7, it is equivalent to compute $\Phi_{\rho_0, \nabla J(\text{End}_{\rho_0}(u))}$. Eq. (20) implies that

$$\begin{aligned} &\Phi_{\rho_0, \nabla J(\text{End}_{\rho_0}(u))} \\ &= \langle \nabla J(\rho(T)), U(T)[U(t)^\dagger H_1 U(t), \rho_0]U(T)^\dagger \rangle_{\rho(T)} \\ &= \langle [\rho_0, U^\dagger(T)[\rho(T), \theta U(T)], [U(t)^\dagger H_1 U(t), \rho_0] \rangle_{\rho_0} \\ &= -\langle \text{ad}_{\rho_0}[\rho_0, U^\dagger(T)\theta U(T)], \text{ad}_{\rho_0}U(t)^\dagger H_1 U(t) \rangle_{\rho_0}. \end{aligned}$$

We note that $[\rho_0, U^\dagger(T)\theta U(T)] \in \mathfrak{p}$. In fact, we have $[\rho_0, \gamma] \in \mathfrak{p}$ for all $\gamma \in \mathbb{C}^{n \times n}$. Indeed, let $\omega \in \mathfrak{h}$. By definition of \mathfrak{h} , we have $[\rho_0, \omega] = 0$. Since $\text{tr}([\rho_0, \gamma]\omega) = -\text{tr}(\gamma[\rho_0, \omega])$, we get $[\rho_0, \gamma] \in \mathfrak{p}$. Therefore, by the definition of $\langle \cdot, \cdot \rangle_{\rho_0}$, we have $\Phi_{\rho_0, \nabla J(\text{End}_{\rho_0}(u))} = -\text{tr}([\rho_0, U(T)^\dagger \theta U(T)]U^\dagger(t)H_1U(t))$. ■

Proposition III.3. The initial value problem defined by Eq. (28) has a unique solution which is globally defined for all

$s \geq 0$.

Proof of Proposition III.3: The uniqueness and local existence of solution for Eq. (28) is straightforward. If u is not a critical point of \mathcal{J} , since $U(\cdot) \in \text{SU}(n)$, Lemma III.2 implies that there exists a constant $C > 0$ depending on ρ_0 , θ , H_1 , and the final time T such that

$$\|\nabla \mathcal{J}(u)\|_{\mathcal{H}} \leq C, \quad \forall u \in \mathcal{H}. \quad (29)$$

Then, by Cauchy-Schwartz inequality, we have

$$\frac{d\|\Pi(s)\|_{\mathcal{H}}}{ds} = \left(\frac{\Pi(s)}{\|\Pi(s)\|_{\mathcal{H}}}, \frac{d\Pi(s)}{ds} \right)_{\mathcal{H}} \leq C.$$

Finally, Growall inequality implies that

$$\|\Pi(s)\|_{\mathcal{H}} \leq Cs. \quad (30)$$

Therefore, the solution of Eq. (28) is globally defined on $[0, \infty[$. ■

Proposition III.4. Given $u_0 \in \mathcal{H}$ which is not a critical point of \mathcal{J} , the solution of Eq. (28) starting from u_0 converges to a connected component of the set of critical points of \mathcal{J} as $s \rightarrow +\infty$.

Proof of Proposition III.4: If u_0 is not a critical point of \mathcal{J} , then

$$\begin{aligned} \frac{d\mathcal{J}(\Pi(s))}{ds} &= \left(\nabla \mathcal{J}(\Pi(s)), \frac{d\Pi(s)}{ds} \right)_{\mathcal{H}} \\ &= \|\nabla \mathcal{J}(\Pi(s))\|_{\mathcal{H}}^2 > 0. \end{aligned} \quad (31)$$

Since $\mathcal{O}(\rho_0)$ is compact, the real-valued function J defined on $\mathcal{O}(\rho_0)$ is bounded. Therefore, $\mathcal{J} = J \circ \text{End}_{\rho_0}$ is also bounded. (31) implies that $\lim_{s \rightarrow +\infty} \mathcal{J}(\Pi(s))$ exists.

We now show that $\frac{d\mathcal{J}(\Pi(s))}{ds}$ is uniformly continuous. By Corollary II.5, Eq. (29), and Corollary II.10 respectively, $\text{End}_{\rho_0}(\cdot)$, $\Pi(\cdot)$, and $G(\cdot)$ are all Lipschitz functions, they are therefore uniformly continuous. As $\nabla J(\cdot)$ is a continuous function defined on the compact set $\mathcal{O}(\rho_0)$, it is also uniformly continuous. Therefore, $\frac{dJ(\pi(s))}{ds}$ is uniformly continuous as composition of uniformly continuous functions.

Since $\lim_{s \rightarrow +\infty} \mathcal{J}(\pi(s))$ exists and $\frac{d\mathcal{J}(\pi(s))}{ds}$ is uniformly continuous, Barbalat's Lemma implies that

$$\lim_{s \rightarrow +\infty} \frac{d\mathcal{J}(\Pi(s))}{ds} = 0.$$

In other words, $\Pi(\cdot)$ converges to a connected component of the set of critical point of \mathcal{J} . ■

Remark III.1. The above result only guarantees the convergence of $\Pi(\cdot)$ to a set of critical points but does not directly imply the existence of $\lim_{s \rightarrow +\infty} \Pi(s)$. We need further information about the set of critical points of \mathcal{J} .

B. Characterization of critical points

Proposition III.5. A control $u \in \mathcal{H}$ is a critical point of \mathcal{J} if and only if

$$\nabla J(\text{End}_{\rho_0}(u)) \in \text{Kernel}(d\text{End}_{\rho_0}^*(u)), \quad (32)$$

which is equivalent to

$$\nabla J(\text{End}_{\rho_0}(u)) \perp \text{Image}(d\text{End}_{\rho_0}(u)), \quad (33)$$

where the orthogonality symbol \perp is taken with respect to the inner product $\langle \cdot, \cdot \rangle_{\text{End}_{\rho_0}(u)}$. An equivalent condition is that the switching function $\Phi_{\rho_0, \nabla J(\text{End}_{\rho_0}(u))}(\cdot)$ is equal to zero almost everywhere on $[0, T]$.

Proof of Corollary III.5:

It suffices to note that the kernel of $d\text{End}_{\rho_0}^*(u)$ is equal to the orthogonal complement of the image of $d\text{End}_{\rho_0}(u)$ with respect to the inner product $\langle \cdot, \cdot \rangle_{\text{End}_{\rho_0}(u)}$. The last condition comes from Corollary II.7. ■

Proposition III.5 together with Lemma III.2 implies the following more explicit characterization.

Corollary III.6. A control $u \in \mathcal{H}$ is a critical point of \mathcal{J} if and only if

$$\text{tr}([\rho_0, U(T)^\dagger \theta U(T)] U^\dagger(t) H_1 U(t)) = 0, \quad \text{for } t \in [0, T], \quad (34)$$

which is equivalent to

$$\text{tr}([\rho(T), \theta] U(t-T)^\dagger H_1 U(t-T)) = 0, \quad \text{for } t \in [0, T]. \quad (35)$$

The following properties are straightforward.

Corollary III.7. Consider $u \in \mathcal{H}$. If $\text{End}_{\rho_0}(u)$ is a critical point of J , then u is a critical point of \mathcal{J} .

Corollary III.8. If $u \in \mathcal{H}$ is a regular control, then u is a critical point of \mathcal{J} if and only if $\text{End}_{\rho_0}(u)$ is a critical point of J .

For later discussion, we distinguish two types of critical points.

Definition III.1. A control $u \in \mathcal{H}$ is a *kinematic critical point* of \mathcal{J} if $\text{End}_{\rho_0}(u)$ is a critical point of J . All other critical points of \mathcal{J} are called *dynamic or non-kinematic critical point*.

Remark III.2. We note that dynamic critical points are necessarily singular in the sense of Definition II.3 while kinematic critical points can be either regular or singular. In the absence of singular controls in \mathcal{H} , all the critical points of \mathcal{J} are kinematic and regular. We also note that the dynamic critical points are necessarily not solutions for **Problem 1**, see Remark I.4.

C. Analysis in the absence of singular controls

The standing assumption of this section is the following:

(H2) all the controls in \mathcal{H} are regular (Definition II.2).

Although (H2) seems restrictive, it allows us to give a complete analysis of the asymptotic behavior of (27) in accordance

with existing numerical simulation results. The goal of this section is to prove the following result.

Theorem III.9. Under (H1) and (H2), every solution of the gradient flow (27) converges to a critical point of \mathcal{J} as $s \rightarrow \infty$. Moreover, for almost all initial conditions, the solution of (27) converges to a solution of **Problem 1**.

We start by giving a more precise characterization of the set of critical points of \mathcal{J} under (H2). For ease of notation, the kernel of $d\text{End}_{\rho_0}(u)$ and the image of $d\text{End}_{\rho_0}^*(u)$ will respectively be denoted by K_u and \mathcal{I}_u .

Proposition III.10. For $i = 1, \dots, M$, let $\mathcal{H}_i := \{u \in \mathcal{H}, \text{End}_{\rho_0}(u) = \rho_i\}$. Under (H2), we have

- (i) the set of critical points of \mathcal{J} is the disjoint union of \mathcal{H}_i with $i = 1, \dots, M$;
- (ii) \mathcal{H}_i 's are submanifolds in \mathcal{H} of co-dimension N ;
- (iii) The tangent space to \mathcal{H}_i at $u \in \mathcal{H}_i$ denoted by $T_u\mathcal{H}_i$ is equal to K_u .

Proof of Proposition III.10: (i) is a consequence of III.8 and the fact that J has only isolated critical points. For all $u \in \mathcal{H}$, $d\text{End}_{\rho_0}(u)$ has finite rank, thus its kernel splits. By (H2), $\text{End}_{\rho_0}(\cdot)$ is a submersion from \mathcal{H} to $\mathcal{O}(\rho_0)$ (cf. [19, Prop. 2.3, p 29]). Therefore, by the Submersion Theorem, the set $\text{End}_{\rho_0}^{-1}(\rho_i)$ is a submanifold in \mathcal{H} of co-dimension N , and $T_u\mathcal{H}_i = K_u$ (cf. [1, Th. 3.5.4, p 175]). ■

By computing the second order Taylor expansion of \mathcal{J} , the following result holds true.

Lemma III.11. For $u \in \mathcal{H}_i$, the Hessian of \mathcal{J} at u is given by

$$\mathcal{A}(u) := d\text{End}_{\rho_0}^*(u) \nabla^2 J(\rho_i) d\text{End}_{\rho_0}(u). \quad (36)$$

Proposition III.12. For $u \in \mathcal{H}_i$, we have

- (i) the kernel of $\mathcal{A}(u)$ is equal to K_u ;
- (ii) the number of positive (resp. negative) eigenvalues of $\mathcal{A}(u)$ is equal to the number of positive (resp. negative) eigenvalues of $\nabla^2 J(\rho_i)$.

Proof of Proposition III.12: For (i), let $v \in \mathcal{H}$. Since $d\text{End}_{\rho_0}^*(u)$ is injective, $\mathcal{A}(u)v = 0$ implies $\nabla^2 J(\rho_i) d\text{End}_{\rho_0}(u)v = 0$. By Lemma II.3, one gets $v \in K_u$. The converse is clear. For (ii), we first note that by (H2) and Lemma II.3, the image of $\mathcal{A}(u)$ is equal to \mathcal{I}_u . Let $g(u)$ be the positive definite symmetric matrix such that $g^2(u) = G(u)$ (see Definition II.6). We set

$$a(\rho_i) := g(u) \nabla^2 J(\rho_i) g(u).$$

Since $g(u) = g^T(u)$, by Sylvester's law of inertia, $a(\rho_i)$ and $\nabla^2 J(\rho_i)$ have the same numbers of positive and negative eigenvalues. Let $\{\nu_k\}_{k=1, \dots, M}$ be the set of eigenvalues of $a(\rho_i)$ and $\{\mu_k\}_{k=1, \dots, M}$ be the corresponding set of orthonormal eigenvectors. For $k = 1, \dots, M$, let

$$v_k := d\text{End}_{\rho_0}^*(u) g(u)^{-1} \mu_k.$$

Then, it is clear that the set $\{v_k\}_{k=1, \dots, M}$ forms a basis of \mathcal{I}_u . Moreover, we have, for $k = 1, \dots, M$,

$$\begin{aligned} \mathcal{A}(u)v_k &= d\text{End}_{\rho_0}^*(u) \nabla^2 J(\rho_i) G(u) g^{-1}(u) \mu_k \\ &= d\text{End}_{\rho_0}^*(u) \nabla^2 J(\rho_i) g(u) \mu_k \\ &= \nu_k d\text{End}_{\rho_0}^*(u) g^{-1}(u) \mu_k \\ &= \nu_k v_k. \end{aligned}$$

Therefore, the non-zero part in the spectrum of $\mathcal{A}(u)$ is equal to the spectrum of $a(\rho_i)$. We conclude that $\nabla^2 J(\rho_i)$ and the restriction of $\mathcal{A}(u)$ to \mathcal{I}_u have the same signature. ■

As a direct consequence of Lemma II.3 and Proposition III.12, we have the following result, which, together with Proposition III.12, will play a crucial role in the convergence analysis of (27).

Corollary III.13.

- (i) For $u \in \mathcal{H}_1$, $\mathcal{A}(u)$ restricted to \mathcal{I}_u is positive definite;
- (ii) For $u \in \mathcal{H}_M$, $\mathcal{A}(u)$ restricted to \mathcal{I}_u is negative definite;
- (iii) For $u \in \mathcal{H}_i$ with $i = 2, \dots, M-1$, $\mathcal{A}(u)$ restricted to \mathcal{I}_u is not definite.

Based on (i) of Proposition III.12, the following result is a generalization of the classical Morse Lemma (see for example [19, Ch. 7, Th. 5.1]). For functions defined on a finite dimensional manifold, a result similar to Proposition III.14 is known as Morse-Bott Lemma. In fact, the following result deals with the case where critical submanifolds are of infinite dimension. For the sake of completeness, a proof will be given in Appendix.

Proposition III.14. Let \mathcal{C} be a connected component of \mathcal{H}_i and $u_c \in \mathcal{C}$. Then, there exist an open neighborhood U of u_c in \mathcal{H} and a smooth chart $\phi : U \mapsto \mathcal{H} = \mathcal{P}_1 \oplus \mathcal{P}_2 \oplus \mathcal{K}$ such that

- (i) the dimensions of \mathcal{P}_1 and \mathcal{P}_2 are equal to N_1^i and $N - N_1^i$ respectively, where N_1^i is the Morse index of u_c ;
- (ii) $\phi(u_c) = 0$, and

$$\phi(U \cap \mathcal{C}) = \{(v_1, v_2, w) \in \mathcal{P}_1 \times \mathcal{P}_2 \times \mathcal{K}, v_1 = v_2 = 0\};$$

- (iii) $\mathcal{J} \circ \phi^{-1}(v_1, v_2, w) = \mathcal{J}(u_c) - \|v_1\|_{\mathcal{H}}^2 + \|v_2\|_{\mathcal{H}}^2$.

Remark III.3. By Corollary III.13, $N_1^1 = 0$, $0 < N_1^i < N$, for $i = 2, \dots, M-1$, and $N_1^M = N$.

Remark III.4. The gradient flow defined by \mathcal{J} and the one defined by $\tilde{\mathcal{J}} := \mathcal{J} \circ \phi^{-1}$ are equivalent in the sense of [2, Theorem, Ch. 1, Sec. 5.3]. The diffeomorphism ϕ provides us with a suitable change of coordinates and allows us to simplify the expression of (27).

Once we have Proposition III.14, the proof of Theorem III.9 is a straightforward adaptation of the proof sketch of [13, Prop. 3.6, Ch. 1, p 20].

Proof of Theorem III.9: We know from Proposition III.4 that the flow converges to a connected component \mathcal{C} of \mathcal{H}_i for some $i \in \{1, \dots, M\}$. Fix an arbitrary $u_c \in \mathcal{C}$ and consider a neighborhood U of u_c small enough. Without loss

of generality, if $\Pi(\cdot)$ is the solution of (27), we can assume that $\Pi(s_0) \in U$ for some $s_0 > 0$ large enough. Using the change of coordinates introduced in Proposition III.14 and taking into account Remark III.4, the gradient flow of \mathcal{J} starting from $\Pi(s_0)$ is equivalent to the gradient flow of $\mathcal{J} \circ \phi^{-1}$ in a neighborhood of \mathcal{C} ,

$$\begin{aligned}\dot{v}_1 &= -v_1, \\ \dot{v}_2 &= v_2, \\ \dot{w} &= 0,\end{aligned}\tag{37}$$

where $(v_1, v_2, w) \in \mathcal{P}_1 \times \mathcal{P}_2 \times \mathcal{K}$ and $(v_1(0), v_2(0), w(0)) = \phi(\Pi(s_0))$. The solution of (37) will be denoted by $\tilde{\Pi}(\cdot)$. Two situations can happen:

- (i) if $\phi(\Pi(s_0)) = (v_{1,0}, 0, w_0)$ for some $v_{1,0} \in \mathcal{P}_1$ and $w_0 \in \mathcal{K}$, then $\lim_{s \rightarrow \infty} \tilde{\Pi}(s) = (0, 0, w_0)$, which implies that

$$\lim_{s \rightarrow \infty} \Pi(s) = \phi^{-1}(0, 0, w_0) \in \mathcal{H}_i;$$

- (ii) if $\phi(\Pi(s_0)) = (v_{1,0}, v_{2,0}, w_0)$ with $v_{2,0} \neq 0 \in \mathcal{P}_2$, then

$$\lim_{s \rightarrow \infty} \tilde{\Pi}(s) = +\infty.\tag{38}$$

This case requires that \mathcal{P}_2 be a subspace of dimension greater than 1, i.e., \mathcal{C} is a connected component of \mathcal{H}_i for some $i \in \{1, \dots, M-1\}$, see Remark III.3. However, if this case happens, (38) implies that $\Pi(\cdot)$ does not converge to \mathcal{C} . Therefore, \mathcal{C} is necessarily a connected component of \mathcal{H}_M . In this case, the flow (37) is reduced to the following

$$\begin{aligned}\dot{v}_1 &= -v_1, \\ \dot{w} &= 0,\end{aligned}\tag{39}$$

where $(v_1, w) \in \mathcal{P}_1 \times \mathcal{K}$ with $\dim \mathcal{P}_1 = N$. The asymptotic behavior of (39) implies that $\Pi(s)$ will converge to an element of $\mathcal{C} \subset \mathcal{H}_M$ as $s \rightarrow \infty$.

We conclude from (i) and (ii) that all the solutions of the gradient flow of \mathcal{J} always converge pointwise. It is also clear that for almost all initial conditions, case (ii) happens, i.e., almost all the solutions converge to a maximum of \mathcal{J} . ■

D. Comments on the role of singular controls

We give in this section a more explicit characterization of singular controls for (1) and then explain some difficulty in the analysis of the asymptotic behavior of (27) related to their presence in \mathcal{H} . We first recall the following result which is a direct application of [5, Th. 6, p 41] or [27, Prop. 5.3.4, p 94] to (1).

Lemma III.15. Let $u \in \mathcal{H}$ and $\rho(\cdot)$ be the corresponding trajectory. Then, u is a singular control if and only if there exists an absolutely continuous application $q : [0, T] \mapsto T\mathcal{O}(\rho_0) \setminus \{0\}$ such that

$$\dot{q}(t) = [H_0 + u(t)H_1, q(t)],\tag{40}$$

and

$$\langle q(t), [H_1, \rho(t)] \rangle_{\rho(t)} = 0, \quad \text{for } t \in [0, T].\tag{41}$$

Using Definition II.1 and (11), we have the following equivalent characterization of singular controls.

Corollary III.16. A control $u \in \mathcal{H}$ is singular if and only if there exists $\Omega_0 \in \mathfrak{p} \setminus \{0\}$ such that

$$\text{tr}(\Omega_0 U^\dagger(t)H_1U(t)) = 0, \quad \text{for } t \in [0, T],\tag{42}$$

with $U(\cdot)$ satisfying

$$\begin{cases} \dot{U}(t) &= (H_0 + u(t)H_1)U(t), & t \in [0, 1], \\ U(0) &= \text{Id}. \end{cases}\tag{43}$$

Proof of Corollary III.16: Assume there exists $\Omega_0 \in \mathfrak{p} \setminus \{0\}$ such that Eqs. (42) and (43) are satisfied. Let $q(\cdot)$ be the solution of Eq. (40) starting from $q(0) := [\rho_0, \Omega_0]$. Then, we have

$$\begin{aligned}q(t) &= U(t)[\rho_0, \Omega_0]U^\dagger(t) \\ &= \rho(t)U(t)\Omega_0U^\dagger(t) - U(t)\Omega_0U^\dagger(t)\rho(t) \\ &= \text{ad}_{\rho(t)}\text{Ad}_{U(t)}\Omega_0.\end{aligned}$$

This implies

$$\begin{aligned}\langle q(t), [H_1, \rho(t)] \rangle_{\rho(t)} &= -\langle \text{ad}_{\rho(t)}\text{Ad}_{U(t)}\Omega_0, \text{ad}_{\rho(t)}\text{Ad}_{U(t)}U^\dagger(t)H_1U(t) \rangle_{\rho(t)} \\ &= -\text{tr}(\Omega_0 (U^\dagger(t)H_1U(t))^\rho) \\ &= -\text{tr}(\Omega_0 U^\dagger(t)H_1U(t)) = 0.\end{aligned}$$

By Proposition III.15, u is singular. The converse is immediate. ■

Remark III.5. This result states that a control u is singular if and only if the real and imaginary parts of the matrix elements of the projection of $U^\dagger(t)H_1U(t)$ on \mathfrak{p} , where $U(\cdot)$ satisfies (43), are \mathbb{R} -linearly dependent functions of t over the time interval $[0, T]$.

We note that, according to Corollary III.7, the elements of \mathcal{H}_i , for $i = 1, \dots, M$, are still critical points of \mathcal{J} called *kinematic* critical points. However, due to possible rank deficiency of the end-point map, the Submersion Theorem may no longer be used and \mathcal{H}_i may not necessarily be submanifolds of \mathcal{H} . More importantly, for $u \in \mathcal{H}_i$, although the expression of the Hessian of \mathcal{J} at u given by (36) is still valid, the two crucial results given in Proposition III.12 may fail. In other words, the non-zero part of the signature of $\mathcal{A}(u)$ may no longer be determined by the signature of $\nabla^2 J(\text{End}_{\rho_0}(u))$. This is the first complication in the asymptotic analysis due to the presence of singular controls.

The second difficulty is the occurrence of *non-kinematic* critical points of \mathcal{J} . We know from Remark I.4 that these critical points are not global maxima of \mathcal{J} . However, nothing *a priori* prevents them from being local maxima and then “attracting” solutions of (27). Although this situation has never been observed in numerical simulations, a formal proof is still missing. A complete spectral analysis on the Hessian of \mathcal{J} needs to be performed. We note that if u is a non-kinematic critical points of \mathcal{J} , the Hessian form of \mathcal{J} at u is given by

$$\begin{aligned}\nabla^2 \mathcal{J}(u)(v, v) &= (v, \mathcal{A}(u)v)_{\mathcal{H}} + \langle \nabla J(\rho), d^2 \text{End}_{\rho_0}(u)(v, v) \rangle_{\rho},\end{aligned}\tag{44}$$

where $v \in \mathcal{H}$ and $\rho := \text{End}_{\rho_0}(u)$.

IV. CONCLUSION

We presented in this paper a gradient-type dynamical system to solve the problem of maximizing quantum observables (**Problem 1**). Under the regularity assumption on the controls (**H2**), we proved that for almost all initial conditions, Eq. (27) converges to a solution of **Problem 1**. We also detailed difficulties related to the presence of singular controls, which constitute the starting point for further investigations. From our point of view, one first needs more explicit characterization of singular controls, then deduces information on the “size” of the set of singular controls \mathcal{S} in the entire control space $L^2([0, T], \mathbb{R})$. The next step is to investigate the optimality status of a “generic” elements of \mathcal{S} . Finally, let us also emphasize that upon due care to numerical details, simulations for extensive systems always achieved the global maximum.

APPENDIX

PROOF OF PROPOSITION III.14

We first note that Morse-Bott Lemma are often stated without proof as a direct consequence of Morse Lemma. A complete proof of this result for functions defined in finite dimensional vector spaces can be found in [3]. We will see in the following that dealing with infinite dimensional critical submanifolds presents no difficulty.

Proof of Proposition III.14: Let \mathcal{C} be a connected component of \mathcal{H}_i and $u_c \in \mathcal{C}$. Since \mathcal{H}_i is a submanifold of \mathcal{H} of co-dimension N , there exist a neighborhood U of u_c in \mathcal{H} and a smooth chart $\varphi : U \mapsto \mathcal{H} = \mathcal{P} \oplus^\perp \mathcal{K}$ such that

- the dimension of \mathcal{P} is equal to N ;
- $\varphi(u_c) = 0$, and $\varphi(U \cap \mathcal{C}) = \{0\} \times \mathcal{K}$.

Fix $w \in \mathcal{K}$ in a neighborhood of 0. Proposition III.12 implies that the Hessian at 0 of the new functional \mathcal{J}_w defined by

$$\mathcal{J}_w(v) := \mathcal{J} \circ \varphi^{-1}(v, w)$$

is non degenerate on \mathcal{P} . Note also that the signature of the Hessian of \mathcal{J}_w at 0 is equal to the one of the restriction of $\mathcal{A}(u_c)$ to \mathcal{I}_{u_c} . Applying Morse Lemma (cf. [19, Ch. 7, Th. 5.1]) to \mathcal{J}_w , there exists a smooth change of coordinates ψ_w , $v \mapsto x := \psi_w(v)$, such that

$$\mathcal{J}_w(\psi_w^{-1}(x)) = (Ax, x),$$

where A is a symmetric matrix which has the same signature of the Hessian of \mathcal{J}_w at 0. Note also that ψ_w depends smoothly on w .

Let $\varphi(u) = (\varphi_1(u), \varphi_2(u)) \in \mathcal{P} \times \mathcal{K}$ and ϕ be the new smooth chart for \mathcal{C} in a neighborhood of u_c defined by

$$\phi(u) := (\psi_{\varphi_2(u)}(\varphi_1(u)), \varphi_2(u)).$$

Then, by construction, if $x = \phi(u)$, we have

$$\mathcal{J} \circ \phi^{-1}(x) = (Ax, x). \quad (45)$$

Proposition III.14 follows from (45). ■

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